Environ Monit Assess (2011) 183:23–39
DOI 10.1007/s10661-011-1903-3

Assessment of Cu, Pb, and Zn contamination in sediment of north western Peninsular Malaysia by using sediment quality values and different geochemical indices

C. K. Yap · B. H. Pang

Received: 5 May 2010 / Accepted: 27 January 2011 / Published online: 23 February 2011

Abstract Surface sediments were collected from the north western aquatic area (13 intertidal sites and 5 river drainages) of Peninsular Malaysia, which were suspected to have received different anthropogenic sources. These sites included town areas, ports, fishing village, industrial areas, highway sides, jetties and some relatively unpolluted sites. The present study revealed that 4.79–32.91 μg/g dry weight for Cu, 15.85–61.56 μg/g dry weight for Pb, and 33.6–317.4 μg/g dry weight for Zn based on 13 intertidal surface sediments while those based on 5 river drainage surface sediments were 10.24–119.6 μg/g dry weight for Cu, 26.7–125.7 μg/g dry weight for Pb and 88.7–484.1 μg/g dry weight for Zn. In general, the metal levels in the drainage sediments are higher than in the intertidal sediments, suggesting dilution factor in the intertidal sediment and direct effluent from point sources in the drainage sediment. In particular, the total concentrations of Cu, Pb, and Zn for the sampling site at Kuala Kurau Town exceeded the Effect Range Median values for Cu, Pb, and Zn for assessments of sediment quality values for freshwater sediment as proposed by MacDonald et al. (Arch Environ Contam Toxicol 39:20–31, 2000), thus adverse biological effects would be observed above this level. Assessment using enrichment factor (using Fe as a normalizer) and geoaccumulation index showed that the three metals at Kuala Kurau Town and Juru Industry drainage were evidenced as having more enrichment and mostly due to non-natural sources. However, caution should be exercised that the interpretation can only become valid when the ratios, indices, and sediment quality values are combined. This is due to the fact that not all the established indices are applicable and, to a certain extent, some of them should be further revised and improved to suit a different metal for Malaysian sediment. Undoubtedly, sites near drainages at Kuala Kurau Town and Juru River Basin need greater attention to mitigate the heavy metal pollution in the future.

Keywords Heavy metals · Peninsular Malaysia · Surface sediment · Geochemical indices

Introduction

Monitoring heavy metal pollution by using sediment in the aquatic ecosystem has been conducted in Malaysia since decades ago due to the fact that quite a number of such studies had been reported

C. K. Yap (✉) · B. H. Pang
Department of Biology, Faculty of Science,
Universiti Putra Malaysia, 43400 Serdang,
Selangor, Malaysia
e-mail: yapckong@hotmail.com

in the literature (Ismail 1993; Ismail et al. 1993, 2004; Yap et al. 2002a, b, 2003, 2006a, b, 2007a, b, 2008a, b, c, 2009). However, these metal data are reported in the literature without comparing with any specific geochemical indices such as enrichment factor (EF) and geoaccumulation index (Igeo). EF and Igeo can be used to assess the extent of sediment contamination.

Studies on heavy metal contamination of sediments often rely on the analysis of total metal concentrations; however, this is not sufficient enough for an understanding of their environmental behaviors since only a fraction of the total metal is available for biological processes (Morillo et al. 2004; Ramirez et al. 2005). According to Zhang et al. (2009), metal contamination cannot be simply evaluated by examining metal concentrations alone. Therefore, it is needed to study the geochemical speciation and distribution of heavy metals in sediments. This is due to the fact that it is difficult to distinguish the natural originated metals from anthropogenic sources in the sediment. Sequential extraction technique is one of the methods to estimate the heavy metal concentrations in the sediments which are contributed from natural or anthropogenic sources. Heavy metal speciation studies are important because slight changes in metal availability and in the environmental conditions can change the toxicity of metals to animals and plants (Gismera et al. 2004).

The geochemical normalization has been used extensively to obtain enrichment factor and to assess anthropogenic contributions of metals in sediments being studied (Acevedo-Figueroa et al. 2006). In the calculation of EF, usually Al or Fe are used as normalizers. In this study, we did not analyze Al concentration in the sediments. Instead, we used Fe to calculate the EF. The reason is that besides Al being one of the most abundant elements on the earth, Fe is also an abundant element in the structure of clay minerals and is also associated with particle surfaces as oxide coatings. In this study, the geochemical normalization was obtained using Fe as the reference element. This is due to the fact that Fe in the estuarine sediment is mainly from natural weathering processes and has been broadly used to normalize the metal concentrations by a lot of researchers (Feng et al. 1998; Schi and Weisberg 1999; Mucha et al. 2003) in order to reduce particle grain size influence because variations in Fe concentration could be explained by particle grain size differences, with fine-grained sediments having high Fe concentrations; besides, its geochemistry is similar to that of many trace metals and its natural sediment concentration tends to be uniform (Daskalakis and O'Connor 1995; Feng et al. 1998). Moreover, Shen (1992) found a significant linear correlation ($R = 0.799$) between Al_2O_3 and Fe_2O_3 was found in Xiamen Harbor surface sediments. Although there is no such study reported from Malaysia, at least there is ground for us to use Fe as a normalizer in this study. Therefore, we believe it is reasonable to use Fe to calculate metal EF.

Table 1 The assessment criteria for enrichment factor (EF) used in this study

References	EF	Degree of enrichment
Zhang and Liu (2002)	$0.5 \leq EF \leq 1.5$	Trace metals may be entirely from crustal materials or natural weathering processes
	$EF > 1.5$	A significant portion of trace metal is delivered from non-crustal materials or non-natural weathering processes
Han et al. (2006)	$EF \leq 2$	Deficiency to minimal metal enrichment
	$EF > 2$	Various degrees of metal enrichment
Acevedo-Figueroa et al. (2006)	$EF < 1$	No enrichment
	$EF < 3$	Minor enrichment
	EF = 3–5	Moderate enrichment
	EF = 5–10	Moderately severe enrichment
	EF = 10–25	Severe enrichment
	EF = 25–50	Very severe enrichment
	EF >50	Extremely severe enrichment

Table 2 Geoaccumulation index (Igeo) in relation to pollution extent according to Müller (1981)

Igeo values	Igeo class	Pollution intensity
>5	6	Very strongly polluted
4–5	5	Strongly to very strongly polluted
3–4	4	Strongly polluted
2–3	3	Moderately to strongly polluted
1–2	2	Moderately polluted
0–1	1	Unpolluted to moderately polluted
<0	0	Unpolluted

According to a review by Zhang et al. (2009), this EF approach has been widely used to study the sources and contamination of trace metals in riverine, estuarine, and coastal environments. It can indicate whether the metals are from natural weathering processes of rocks or from anthropogenic sources and reflect the status of environmental contamination. The assessment criteria for EF used in this study followed those suggested by Zhang and Liu (2002), Han et al. (2006) and Acevedo-Figueroa et al. (2006). The degrees of enrichments are given in Table 1.

Another criterion to evaluate the heavy metal pollution in the surface sediments is the Igeo by Müller (1981). This index is to determine and define metals contamination in sediments, by comparing current concentrations with pre-industrial levels. The Igeo index can assess the estimation of these pollution process. Müller (1981) has distinguished seven classes of geoaccumulation indexes (Table 2).

The objective of this study was to assess the contamination of Cu, Pb, and Zn collected from north western of Peninsular Malaysia by comparing to sediment quality values (SQVs), EF and Igeo values besides the ratios of nonresistant to resistant geochemical fractions.

Materials and methods

Sediment samples were collected in 18–20 April 2005 from 18 sampling sites from the north western intertidal area (13 sites) and drainage (5 sites) of Peninsular Malaysia (Fig. 1). The positions, sampling dates and site descriptions are given in Table 3. The top 3–5 cm of surface sediments were collected at each sampling site. Each sediment sample was put in a plastic bag and frozen prior to analysis. The salinity of the surface water (0–20 cm) samples recorded directly in the field at each sampling station were salinity by using a Hydrolab Datasonae 4a water quality multi-probe.

Sediment samples were dried by using an oven at 60°C until constant dry weights. Later, the dried sediments were pounded by using a clean pestle and mortar and were sieved through a 63-μm stainless steel aperture. While sifting, the sieve was shaken vigorously to produce homogeneity (Yap et al. 2002a) and stored in clean and new plastic bags.

The direct aqua-regia method was used to determine the concentrations of Cu, Pb, and Zn in the dried sediment samples. Firstly, about 1 g of each dried sample was weighed and digested in a combination of concentrated nitric acid (HNO_3, AnalaR grade, BDH 69%) and perchloric acid ($HClO_4$, AnalaR grade, BDH 60%) in the ratio of 4:1. After that, the tubes were put into the digestion block at the low temperature (40°C) for 1 h and then the temperature was increased to 140°C for at least 3 h. The digested samples were diluted to 40 ml by double distilled water and filtered through Whatman No.1 (filter speed: medium) filter paper in a funnel into acid-washed pillboxes. They were stored until metal determination.

Geochemical fractions of Cu, Pb, and Zn in the sediment were obtained by using the sequential extraction technique which was described by Badri and Aston (1983) and modified by Yap et al. (2002a). They are four fractions considered in this method. These four fractions, extraction solutions and conditions employed by each fraction were:

1. Easily, freely, leacheable, or exchangeable (EFLE): about 10 g of the sample was shaken continuously with 50 ml of 1.0 M ammonium acetate (NH_4CH_3COO), at pH 7.0 and at room temperature pH 7.0 for 3 h.
2. Acid-reducible: the residue from the first step was shaken continuously with 50 ml of 0.25 M hydroxylammonium chloride ($NH_2OH{\cdot}HCl$)

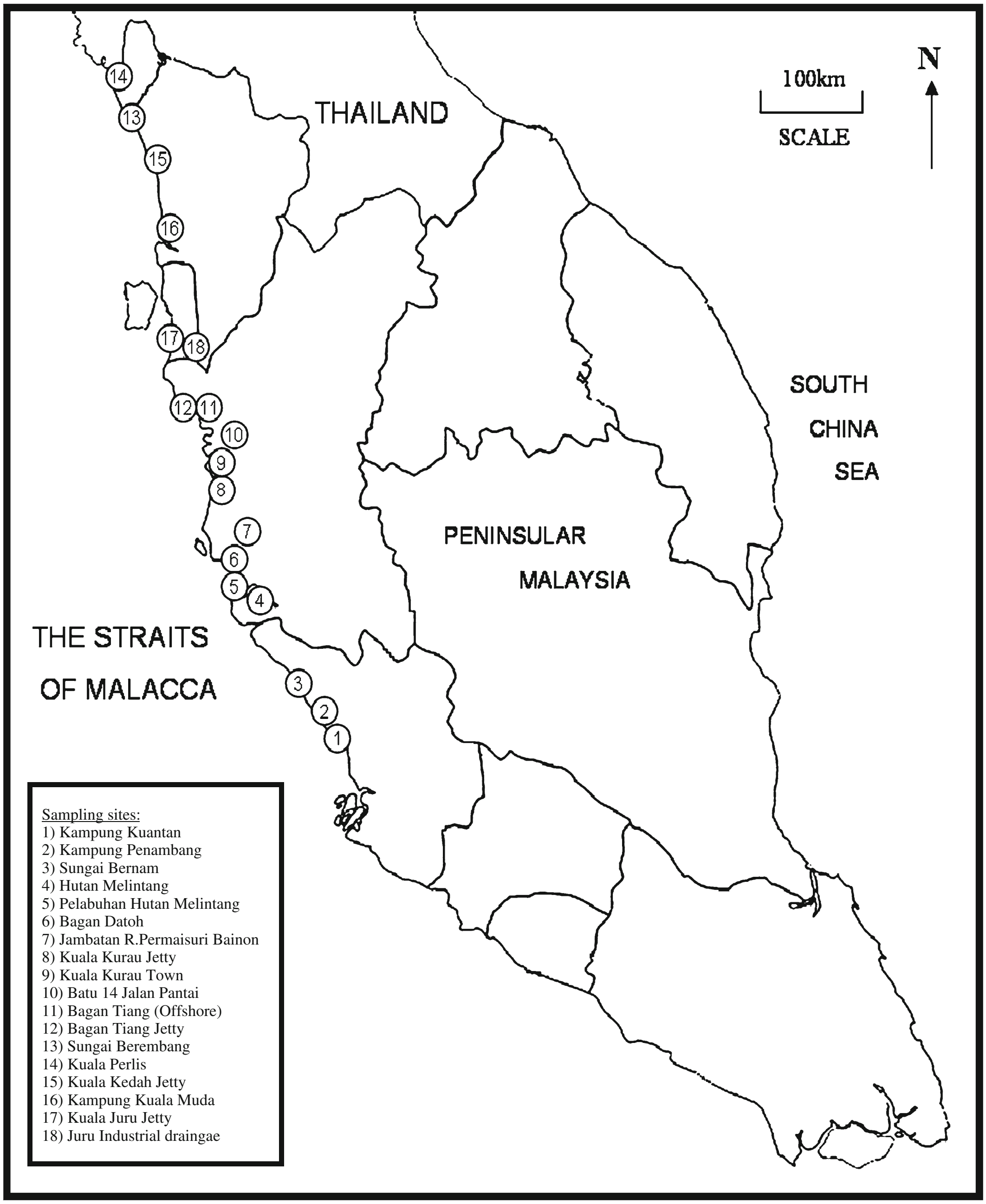

Fig. 1 Sampling locations of surface sediments in the north western coast of Peninsular Malaysia

that had been acidified with HCl to pH 2, at room temperature for 3 h.

3. Oxidisable-organic: the residue from the second step was first oxidized with 30% H_2O_2 in a water bath at 90–95°C. After being oxidized, the residue was shaken continuously with 50 ml of 1.0 M ammonium acetate (NH_4CH_3COO) that had been acidified with HCl to pH 2, at room temperature for 3 h.
4. Resistant: the residue from the third step was digested with 10 ml of an aqua-regia solution that comprised of a combination of nitric acid

Table 3 Positions, sampling dates and description of sampling sites for the intertidal and drainage sediment collected from the north western part of Peninsular Malaysia

St	Location	Latitude	Longitude	Date	Site description	Water salinity	Type
1	Kampung Kuantan, Selangor	101°18.093′ E	03°21.745′ N	18 Apr 2005	A riverside with recreational park	0.04	Freshwater
2	Kampung Penambang, Selangor	101°18.096′ E	03°21.744′ N	18 Apr 2005	A small jetty and a fishing village	6.33	Marine
3	Sungai Bernam, Selangor	101°14.965′ E	03°21.599′ N	18 Apr 2005	Under a bridge of a main highway	0.33	Freshwater
4	Hutan Melintang, Perak	100°55.965′ E	03°52.345′ N	18 Apr 2005	A fishing village	6.06	Marine
5	Pelabuhan Hutan Melintang, Perak	101°14.965′ E	03°21.599′ N	18 Apr 2005	An abandoned port, fishing village in the other side	8.48	Marine
6	Bagan Datoh, Perak	100°47.150′ E	03°59.563′ N	18 Apr 2005	A fishing village	4.98	Marine
7	JambatanRajaPermaisuri Bainon, Perak	100°39.335′ E	04^{o}16.803′ N	18 Apr 2005	An industrial area	28.96	Marine
8	Kuala Kurau Jetty, Perak	100°25.867′ E	05°00.928′ N	19 Apr 2005	A jetty	13.34	Marine
9	Kuala Kurau Town, Perak	100°26.017′ E	05°01.052′ N	19 Apr 2005	A small town with shops	0.21	Freshwater
10	Batu 14 Jalan Pantai, Perak	100°24.779′ E	05°01.106′ N	19 Apr 2005	A highway side	0.16	Freshwater
11	Bagan Tiang (offshore), Perak	100°22.459′ E	05°08.517′ N	19 Apr 2005	An offshore aquacultural site	26.51	Marine
12	Bagan Tiang Jetty, Perak	100°23.840′ E	05°06.702′ N	19 Apr 2005	A jetty and fishing village	3.81	Marine
13	Sungai Berembang, Kedah	100°08.787′ E	06°21.313′ N	19 Apr 2005	A rocky beach	30.71	Marine
14	Kuala Perlis, Perlis	100°07.740′ E	06°23.927′ N	19 Apr 2005	A busy jetty	28.53	Marine
15	Kuala Kedah Jetty, Kedah	100°17.149′ E	06°06.333′ N	20 Apr 2005	A jetty and fishing village	25.43	Marine
16	Kampung Kuala Muda, Kedah	100°21.735′ E	05°34.343′ N	20 Apr 2005	A jetty and fishing village	17.68	Marine
17	Kuala Juru Jetty, Penang	100°24.518′ E	05°20.410′ N	20 Apr 2005	A jetty and fishing village	21.8	Marine
18	Juru Industrial drainage, Kuala Juru, Penang	100°26.011′ E	05°20.105′ N	20 Apr 2005	A industrial area	0.62	Freshwater

(AnalaR grade, BDH 69%) and perchloric acid (AnalaR grade, BDH 60%) in the ratio of 4:1 for 1 h at the temperature of 40°C. The temperature was then increased to 140°C for an additional 3 h.

Before the next fractionation, the residue for each fraction was weighed. Residue was rinsed by 20 ml double distilled water. After that it was filtered through a Whatman No.1 (Filter speed: medium) filter paper in a funnel and the filtrate were stored for the next step. For each fraction of the sequential extraction procedure, a blank was employed by using the same procedure to ensure that the samples and chemicals used were free of contamination.

After filtration, the sample was determined for Cu, Pb, and Zn by using an air–acetylene flame atomic absorption spectrophotometer, an inorganic analytical instrument made by Perkin-Elmer Model AAnalyst 800. All the data were presented in micrograms per gram dry weight basis.

The quality of the method used was checked with a Certified Reference Material for Soil (International Atomic Energy Agency, Soil-5, Vienna, Austria). The agreement between the analytical results for the reference material and its certified values for each metal was satisfactory with the percentages of recovery being between 88.3% for Cu (certified value, 77.1 μg/g; measured value, 68.1 μg/g), 106.4% for Pb (certified value, 129 μg/g; measured value, 137 μg/g), and 87.8% for Zn (certified value, 368 μg/g; measured value, 323 μg/g). Procedural blanks and quality control samples made from the standard solutions for Cu, Pb, and Zn were prepared from 1,000 mg/L stock solution (MERCK Titrisol) of each metal, were analyzed for every five to ten samples in order to check for sample accuracy.

According to Ergin et al. (1991) the metal EF is defined as follows:

$$\mathrm{EF} = \frac{\left(\frac{\mathrm{Me}}{\mathrm{Fe}}\right)\text{Sample}}{\left(\frac{\mathrm{Me}}{\mathrm{Fe}}\right)\text{Background}}$$

where (Me/Fe)Sample is the metal to Fe ratio in the samples of interest; (Me/Fe)Background is the natural background value of metal to Fe ratio. As we do not have metal background values for our study area, we used the values from crust materials [Cu, 32 μg/g; Pb, 16 μg/g; Zn, 127 μg/g; and Fe, 35,900 μg/g] (Martin and Whitfield 1983). The above equation used in the present study can estimate the EF of Cu, Pb and Zn in the sediments of the sampling sites using Fe as a normalizer to correct for differences in sediments grain size and mineralogy.

The Igeo index can be calculated by the following equation:

$$\mathrm{Igeo} = \log 2\left(\frac{Cn}{1.5\,Bn}\right)$$

where Cn is the measured concentration of the examined metal (n) in the sediment and Bn is the geochemical background concentration of the metal (n). Factor 1.5 is the background matrix correction factor due to lithogenic effects. Because we did not have the background values of the metals of interest, same as we did in EF calculation, we adopt the earth crust values (Martin and Whitfield 1983) in Igeo calculation.

Results and discussion

The salinity values recorded in situ on the water surface from 20 sampling sites are given in Table 3. The salinity ranges are 0.04–30.71. From the salinity data, it is clear that Kg. Kuantan (0.04 ppt) and Sg. Bernam (0.33 ppt) are rivers with little influence from the marine seawater while Kuala Kurau Town (0.21 ppt), Batu 14 Jalan Pantai at Kurau (0.16 ppt), and Juru Industrial drainage (0.62 ppt) are concrete drainages potentially receiving effluents from industrial discharges or domestic wastes. Thus, these five sites are considered as freshwater sampling sites. Although these salinity data are difficult to explain the present metal data on the surface sediments, these data can, at least, provide us whether the influence of marine seawater is significant.

The total concentrations of Cu, Pb, and Zn for all sampling sites based on aqua-regia method are presented in Tables 4, 5, and 6, respectively while Table 7 shows the overall results dividing into marine and freshwater sediments. The ranges for total concentrations in the marine sediment were 79–32.91 μg/g dry weight for Cu, 15.85–61.56 μg/g dry weight for Pb, and 33.6–317.4 μg/g dry weight

Table 4 Cu concentrations (μg/g dry weight) of four geochemical fractions and their summation, ratios of nonresistant to resistant fractions (Non*R*/*R*), total concentrations based on direct aqua-regia method (Total), enrichment factor (EF) and geoaccumulation index (Igeo) in the surface sediments collected from 18 sampling sites

No.	Sampling sites	Total	F1	F2	F3	F4	Sum	Non*R*/*R*	Fe	EF	Igeo
1[a]	Kampung Kuantan	11.14	0.14	0.09	3.16	8.65	12.04	0.392	27,723.75	0.451	−2.107
2	Kampung Penambang	4.79	0.15	0.09	1.41	9.19	10.84	0.180	16,181.87	0.332	−3.325
3[a]	Sungai Bernam	10.34	0.09	0.05	1.21	9.72	11.08	0.139	29,068.05	0.399	−2.215
4	Hutan Melintang	13.17	0.07	0.04	2.95	10.16	13.21	0.301	25,116.62	0.589	−1.866
5	Pelabuhan Hutan Melintang	8.32	0.09	0.12	1.9	10.89	13.00	0.194	23,977.88	0.389	−2.528
6	Bagan Datoh	14.20	0.17	0.12	3.28	14.99	18.58	0.238	32,403.23	0.492	−1.757
7	J.Raja Permaisuri Bainon	19.24	0.42	0.18	19.88	8.02	28.50	2.554	15,425.63	1.400	−1.319
8	Kuala Kurau Jetty	12.91	0.2	0.09	4.64	11.55	16.47	0.427	35,365.01	0.410	−1.895
9[a]	Kuala Kurau town	119.64	0.3	0.17	67.01	60.43	127.91	1.117	50,172.08	2.676	1.318
10[a]	Batu 14 Jalan Pantai	10.24	0.24	0.09	2.25	11.94	14.51	0.216	38,403.14	0.299	−2.229
11	Bagan Tiang (offshore)	13.58	0.26	0.19	5.86	14.3	20.60	0.441	30,468.75	0.500	−1.822
12	Bagan Tiang Jetty	9.79	0.46	0.22	2.78	10.97	14.43	0.315	27,258.92	0.403	−2.294
13	Sungai Berembang	5.26	0.2	0.25	3.29	6.12	9.86	0.611	30,814.39	0.192	−3.190
14	Kuala Perlis	21.25	0.21	0.1	5.46	15.04	20.81	0.384	24,274.39	0.983	−1.176
15	Kuala Kedah Jetty	15.67	0.24	0.02	2.58	19.28	22.12	0.147	25,580.93	0.688	−1.615
16	Kuala Muda	20.38	0.1	0.04	9.22	20.1	29.45	0.466	19,212.29	1.191	−1.236
17	Kuala Juru Jetty	32.91	0.52	0.14	18.6	26.27	45.54	0.733	27,595.17	1.338	−0.545
18[a]	Juru industrial drainage	60.92	0.15	0.05	49.91	34.61	84.71	1.448	40,790.31	1.676	0.344

[a] Surface sediments collected from rivers or drainages

*F*1 easily, freely, leachable or exchangeable; *F*2 acid-reducible; *F*3 oxidisable-organic; *F*4 resistant., *NonR/R* summation of F1, F2 and F3 fractions divided by F4 fraction

Table 5 Pb concentrations (μg/g dry weight) of four geochemical fractions and their summation, and total concentrations based on direct aqua-regia method (Total), enrichment factor (EF) and geoaccumulation index (Igeo) in the surface sediments collected from 18 sampling sites

No.	Sampling sites	Total	F1	F2	F3	F4	Sum	Non*R*/*R*	Fe	EF	Igeo
1[a]	Kampung Kuantan	26.74	1.52	2.52	19.68	34.71	58.43	0.683	27,723.75	2.163	0.156
2	Kampung Penambang	15.85	3.49	2.87	11.11	30.45	47.92	0.574	16,181.87	2.196	−0.599
3[a]	Sungai Bernam	33.63	3.81	4.06	14.78	44.8	67.45	0.506	29,068.05	2.594	0.487
4	Hutan Melintang	18.13	5.28	5.48	17.58	34.99	63.33	0.810	25,116.62	1.618	−0.405
5	Pelabuhan Hutan Melintang	42.33	3.14	2.15	16.32	40.33	61.94	0.536	23,977.88	3.958	0.819
6	Bagan Datoh	61.56	2.97	3.5	22.74	51.88	81.09	0.563	32,403.23	4.260	1.359
7	J.Raja Permaisuri Bainon	34.33	7.55	6.13	16.07	21.67	51.42	1.373	15,425.63	4.990	0.516
8	Kuala Kurau Jetty	38.10	4.53	5.56	19	32.45	61.54	0.896	35,365.01	2.416	0.667
9[a]	Kuala Kurau Town	125.68	4.73	2.09	80.84	72.44	160.1	1.210	50,172.08	5.617	2.389
10[a]	Batu 14 Jalan Pantai	43.91	3.01	2.34	22.13	53.65	81.13	0.512	38,403.14	2.564	0.872
11	Bagan Tiang (Offshore)	38.12	4.66	5.04	25.77	42.76	78.23	0.830	30,468.75	2.805	0.668
12	Bagan Tiang Jetty	27.58	4.83	3.84	16.75	32.37	57.79	0.785	27,258.92	2.269	0.201
13	Sungai Berembang	31.82	3.46	4.99	24.66	28.63	61.74	1.156	30,814.39	2.315	0.407
14	Kuala Perlis	25.77	3.29	1.1	19.01	27.48	50.88	0.852	24,274.39	2.380	0.103
15	Kuala Kedah Jetty	26.81	3.95	2.06	14.95	37.25	58.21	0.563	25,580.93	2.350	0.160
16	Kuala Muda	31.92	4	2.07	19.7	41.72	67.49	0.618	19,212.29	3.725	0.411
17	Kuala Juru Jetty	30.20	4.58	2.77	26.01	43.01	76.37	0.776	27,595.17	2.454	0.332
18[a]	Juru Industrial drainage	65.32	2.24	1.5	33.78	80.11	117.63	0.468	40,790.31	3.590	1.444

[a] Surface sediments collected from rivers or drainages

F1 easily, freely, leachable or exchangeable; *F2* acid-reducible; *F3* oxidisable-organic; *F4* resistant., *NonR/R* summation of F1, F2 and F3 fractions divided by F4 fraction

Table 6 Zn concentrations (μg/g dry weight) of four geochemical fractions and their summation, and total concentrations based on direct aqua-regia method (Total), enrichment factor (EF) and geoaccumulation index (Igeo) in the surface sediments collected from 18 sampling sites

No.	Sampling sites	Total	F1	F2	F3	F4	Sum	Non*R*/*R*	Fe	EF	Igeo
1[a]	Kampung Kuantan	119.72	1.01	33.39	47.29	71.51	153.21	1.14	27,723.75	1.22	−0.67
2	Kampung Penambang	54.68	0.7	7.99	15.75	49.38	73.83	0.49	16,181.87	0.96	−1.80
3[a]	Sungai Bernam	101.32	0.91	7.18	19.55	95.68	123.32	0.29	29,068.05	0.99	−0.91
4	Hutan Melintang	83.8	0.97	10.82	22.84	71.07	105.7	0.49	25,116.62	0.94	−1.18
5	Pelabuhan Hutan Melintang	71.15	0.37	4.08	13.63	76.21	94.28	0.24	23,977.88	0.84	−1.42
6	Bagan Datoh	87.99	0.81	11.09	20	72.35	104.25	0.44	32,403.23	0.77	−1.11
7	J.Raja Permaisuri Bainon	38.95	6.03	16.15	16.75	17.54	56.47	2.22	15,425.63	0.71	−2.29
8	Kuala Kurau Jetty	74.64	0.77	5.88	24.87	62.03	93.54	0.51	35,365.01	0.60	−1.35
9[a]	Kuala Kurau Town	429.46	51.93	63.35	86.58	127.36	329.21	1.58	50,172.08	2.42	1.17
10[a]	Batu 14 Jalan Pantai	88.74	4.85	20.2	39.14	58.51	122.71	1.10	38,403.14	0.65	−1.10
11	Bagan Tiang (Offshore)	106.04	1.8	4.33	49.26	83.06	138.45	0.67	30,468.75	0.98	−0.85
12	Bagan Tiang Jetty	73.22	1.25	1.92	24.95	71.58	99.7	0.39	27,258.92	0.76	−1.38
13	Sungai Berembang	57.32	2.07	3.52	40.42	32.75	78.75	1.40	30,814.39	0.53	−1.73
14	Kuala Perlis	55.73	1.11	4.03	20.89	44.89	70.91	0.58	24,274.39	0.65	−1.77
15	Kuala Kedah Jetty	53.21	0.76	7.09	13.16	53.13	74.14	0.40	25,580.93	0.59	−1.84
16	Kuala Muda	33.6	3.93	5.56	11.75	28.81	50.06	0.74	19,212.29	0.49	−2.50
17	Kuala Juru Jetty	317.39	24.96	58.02	84.66	136.01	303.65	1.23	27,595.17	3.25	0.74
18[a]	Juru Industrial drainage	484.14	59.37	63.26	84.34	413	619.95	0.50	40,790.31	3.36	1.35

[a] Surface sediments collected from rivers or drainages

F1 easily, freely, leachable or exchangeable; *F2* acid-reducible; *F3* oxidisable-organic; *F4* resistant., *NonR/R* summation of F1, F2 and F3 fractions divided by F4 fraction

Table 7 Comparisons of overall concentrations (μg/g dry weight) of Cu, Pb and Zn from the present study with those established Interim Sediment Quality Values (ISQVs), average shale, average sediment and continental crusts

Metals	Type	Fractions	Mean ± SE
Cu		Aver. shale[a]	45
		Aver. sediment[b]	33
		Continental crust[c]	25
	Marine	15 sites	14.73 ± 2.10 (4.79–32.91)
		ISQV-low[d]	65
		ISQV-high[d]	270
	Freshwater	5 sites	42.46 ± 21.62 (10.24–119.64)
		ERL[e]	70
		ERM[e]	390
Pb		Aver. shale[a]	20
		Aver. sediment[b]	19
		Continental crust[c]	14.8
	Marine	15 sites	32.50 ± 3.20 (15.85–61.56)
		ISQV-low[d]	75
		ISQV-high[d]	218
	Freshwater	5 sites	59.06 ± 17.89 (26.74–125.68)
		ERL[e]	35
		ERM[e]	110
Zn		Aver. shale[a]	95
		Aver. sediment[b]	95
		Continental crust[c]	65
	Marine	Intertidal sediments (15 sites) This study	85.21 ± 20.13 (33.6–317.4)
		ISQV-low[d]	200
		ISQV-high[d]	410
	Freshwater	5 sites	244.7 ± 87.2 (88.74–484.1)
		ERL[e]	120
		ERM[e]	270

Values in brackets indicate minimum-maximum values

[a] Turekian and Wedepohl (1961)

[b] Salomons and Forstner (1984)

[c] Wedepohl (1995)

[d] Chapman et al. (1999)

[e] MacDonald et al. (2000)

for Zn based on 13 intertidal surface sediments while those based on 5 river drainage surface sediments were 10.24–119.6 μg/g dry weight for Cu, 26.7–125.7 μg/g dry weight for Pb, and 88.7–484.1 μg/g dry weight for Zn. Kuala Kurau Town drainage was found to have the elevated concentrations of Cu, Pb, and Zn while Juru Industrial drainage site had relatively higher metal levels compared to other sites. In general, Kuala Juru Jetty and Juru Industrial drainage had elevated levels of Zn.

The total concentrations of Cu, Pb, and Zn in the sediments collected from the north western part of Peninsular Malaysia were compared with those reported from this region and Malaysia (Table 8). For Cu in the marine sediment, comparing with Malaysian studies, the ranges are comparable and within those reported for the intertidal of Peninsular Malaysia and higher than Tg. Piai and Bintulu coast. When it is compared to regional studies, the Cu ranges are lower than Pearl River Delta/Estuary, Yangtze River (intertidal zone) and Western Xiamen Bay (China), Coastal Alang-Sosiya and Mandovy estuary (India), Central Java Coast (Indonesia), Kaoshiung Harbor (Taiwan), and Victoria Harbor (Hong Kong). However, it is higher than mangrove area, Kranji and Tekong Island (Singapore) and Dumai coast (Indonesia).

For Pb marine sediment (Table 8), the present range is lower than Pearl River Delta (China), Coastal Alang-Sosiya (India) and Kaoshiung Harbor (Taiwan) but higher than several areas including Central Java Coast (Indonesia), mangrove area, Kranji and Tekong Island (Singapore), Mandovy estuary (India), and Western Xiamen Bay and Yangtze River (intertidal zone; China). For Zn in the marine sediment (Table 8), comparing with Malaysian studies, the ranges are

Table 8 Comparison of Cu, Pb and Zn concentrations (µg/g dry weight) of surface sediment data reported from Malaysia and this region

No.	Area	Cu	Pb	Zn	References
	Regional studies				
1.	Pearl River Delta, China	8.70–140	48.10–264.2	46.4–533.3	Cheung et al. (2003)
2.	Coastal Alang-Sosiya, India	85.2–313	66–263	718.02–1,483	Reddy et al. (2004)
3.	Semarang, Indonesia	33–72	18–44	17–36	Takarina et al. (2004)
4.	Mangrove area, Singapore	7.06–32.0	12.3–30.9	51–120	Cuong et al. (2005)
5.	Mandovy estuary, India	11.5–77.5	4.5–46.5	19.9–86	Alagarsamy (2006)
6.	Kranji and Tekong Island, Singapore	7.70–17.9	26.1–29.8	49–62	Cuong and Obbard (2006)
7.	Kaoshiung Harbor, Taiwan	5–946	9.5–470	52–1,369	Chen et al. (2007)
8.	Pearl River Estuary, China	8.9–351.2	12.3–86	120–478	Li et al. (2007)
9.	Western Xiamen Bay, China	19–97	45–60	139	Zhang et al. (2007)
10.	Victoria Harbour, Hong Kong	16–280	21–85	52–221	Chloe et al. (2008)
11.	Yangtze River (intertidal zone), China	6.87–49.7	18.3–44.1	47–154	Zhang et al. (2009)
12.	Dumai coast, Indonesia	1.61–13.8	14.63–84.9	31–87	Amin et al. (2009)
	Malaysian studies				
1.	West coast of Peninsular Malaysia	< 6.00	1.00–45.00	50–1,400	Ismail et al. (1993)
2.	Bintulu coastal waters	7.00–13.00	11.00–36.00	39–91	Ismail (1993)
3.	Straits of Johore	4.40 ± 0.90	30.8 ± 4.8	26.1	Mat et al. (1994)
4.	Juru River	14.00–72.00	9.00–78.00	29–316	Lim and Kiu (1995)
5.	Johore Straits	10.8–92.9	26.4–69.9	68–231	Wood et al. (1997)
6.	Offshore west coast of Peninsular Malaysia	0.25–13.8	3.59–25.4	–	Yap et al. (2002b)
7.	Intertidal west coast of Peninsular Malaysia	0.40–315	0.96–69.8	–	Yap et al. (2002b)
8.	Offshore west coast of Peninsular Malaysia	–	–	4.00–79.05	Yap et al. (2003)
9.	Intertidal west coast of Peninsular Malaysia	–	–	3–306	Yap et al. (2003)
10.	Kelana Jaya Lakes, Selangor	7.37–73.6	–	107–529	Ismail et al. (2004)
11.	Tg. Piai, Peninsular Malaysia	3.43–3.81	16.5–21.6	40–43	Yap et al. (2006a)
12.	Sri Serdang Industrial Area	43.0–551.2	56.1–404.3	169–296	Yap et al. (2006b)
13.	Sepang River, Selangor	2.88–161	–	34–421	Yap et al. (2007a)
14.	Polluted drainage sediments from Peninsular Malaysia (6 sites)	8.77–1,019	57.42–1,267	330–484	Yap et al. (2007b)
15.	East coast of Peninsular Malaysia (10 sites)	12.96	–	55–86	Yap et al. (2008a)
16.	South coast of Peninsular Malaysia (5 sites)	38.8	–	38.1–221	Yap et al. (2008a)
17.	West coast of Peninsular Malaysia (5 sites)	31.13	–	36–395	Yap et al. (2008a)
18.	Sri Serdang Industrial Area, Selangor	48.76	23.52	–	Yap et al. (2008b)
19.	Six intertidal area and 4 urban drainage sites, Selangor	6.64–122.7	26.0–227.7	–	Yap et al. (2008b)
20.	Intertidals and drainages, Selangor	–	–	50–336	Yap et al. (2008c)
21.	Sri Serdang Industrial Area, Selangor (1 site)	347.6	–	219	Yap et al. (2009)
22.	Northern part of Peninsular Malaysia (13 marine sediment, 2005)	4.79–32.91	15.85–61.56	33.6–317.4	This study
23.	Northern part of Peninsular Malaysia (5 freshwater sediment, 2005)	10.24–119.6	10.24–125.68	88.7–484.1	This study

comparable and within those reported for the intertidal of Peninsular Malaysia and higher than offshore west coast of Peninsular Malaysia, Tg. Piai, Johore Straits and Bintulu coast. When it is compared to regional studies, the Zn ranges are lower than Pearl River Delta/Estuary (China), Coastal Alang-Sosiya (India), Kaoshiung Harbor (Taiwan). However, it is higher than Yangtze River (intertidal zone) and Western Xiamen Bay (China), mangrove area, Kranji and Tekong Island (Singapore), Mandovy estuary (India) and Semarang and Dumai coasts (Indonesia), and Victoria Harbor (Hong Kong).

For the freshwater sediments, the present ranges for Cu, Pb and Zn are comparable and within those reported from Malaysia such as

Serdang Industrial drainage and Selangor industrial and urban drainages (Table 8).

Although comparisons with other reported data can give us a picture of the overall contamination level in Malaysia, it is still uncertain about the environmental consequences by these metals. Therefore, in order to estimate possible environmental consequences of Cu, Pb, and Zn at the studied sites, the metals are compared to the interim sediment quality values (ISQVs) for Hong Kong of effect range low (ERL) and effect range median (ERM) for marine sediment as proposed Chapman et al. (1999), as shown in Table 7. According to a review done by Praveena et al. (2008), Hong Kong's ISQVs are the most appropriate guidelines that meet the prioritization criteria and consistent with international initiatives and regulations (Chapman et al. 1999). The present results showed that Cu and Pb concentrations in all the sampling sites were below values for ERL for Cu (65 μg/g) and Pb (75 μg/g), indicating relatively "unpolluted" status. However, site at Kuala Juru Jetty exceeded the ERL (200 μg/g) for Zn.

For freshwater SQVs (Table 7), the present data are compared to ERL and ERM as proposed by MacDonald et al. (2000) to assess the ecotoxicology of Cu, Pb, and Zn concentrations in sediments. Only Kuala Kurau Town drainage exceeded the ERL for Cu (70) but lower than the ERM value (390). Four other sites are below the Cu ERL value. For Pb, Batu 14 Jalan Pantai and Juru Industrial drainage exceeded the Pb ERL value (35) and Kuala Kurau Town exceeded the ERM value (110). For Zn, sites at Kuala Kurau Town and Juru Industrial drainage exceeded the ERM (270) while other sites are below ERL. Low-range values (ERLs) are concentrations below which adverse effects upon sediment dwelling fauna would be infrequently expected. In contrast, the ERMs represent chemical concentrations above which adverse effects are likely to occur (MacDonald et al. 2000). Therefore, sediment at Kuala Kurau Town and Juru Industrial drainage might have adverse biological effects since at least one metal exceeded the ERM values.

The present Cu levels are also lower than those for average shale (Turekian and Wedepohl 1961) and average sediment (Salomons and Forstner 1984) for all sampling sites except for Kuala Juru Jetty which had exceeded the continental crust value (Wedepohl 1995). For the freshwater sediments, Kuala Kurau Town and Juru Industrial drainage all exceeded the three values.

For Pb, all the sampling sites for marine or freshwater all exceeded the values for average shale (Turekian and Wedepohl 1961), average sediment (Salomons and Forstner 1984) and continental crust value (Wedepohl 1995). For Zn in the marine sediment, 6 out of 13 sites are below the continental crust value while another 5 sites exceeded the continental crust value. Only Bagan Tiang offshore and Kuala Juru Jetty exceeded all the values for average shale (Turekian and Wedepohl 1961), average sediment (Salomons and Forstner 1984), and continental crust value (Wedepohl 1995). For the Zn in freshwater sediment, all the sampling sites (except for Batu 14 Jalan Pantai only exceeded continental crust value) exceeded all the three values.

In order to know the relative contribution of anthropogenic source, ratios of nonresistant to resistant fractions are established for Cu (Table 4), Pb (Table 5), and Zn (Table 6). From Table 4, it is found that sites at J. Raja Permaisuri Bainon, Kuala Kurau Town, Juru Industrial drainage had major contribution (>50%) of Cu due to nonresistant fraction as the ratios are >1.0. From Table 5, sites at J. Raja Permaisuri Bainon, Kuala Kurau Town, and Sungai Berembang had major contribution (>50%) of Pb. For Zn (Table 6), sites at Kampung Kuantan, J. Raja Permaisuri Bainon, Kuala Kurau Town, Batu 14 Jalan Pantai, Sungai Berembang, Kuala Juru Jetty were all dominated by nonresistant fractions of Zn but not in the site at Juru Industrial drainage. This could be due to the fact that the chemical extractants used to fractionate the first three nonresistant fractions were not strong enough and could not fully extract these geochemical fractions. It is now well reported that a decrease in the relative importance of the resistant fraction was associated with contaminated conditions and this is well documented in the literatures (Ismail et al. 2004; Yap et al. 2007a, 2008a, b).

Either comparison with reported studies or knowledge on the contribution by the anthropogenic sources can little explain the degree of contamination. Therefore, in order to know the metal degrees of contamination, the application of established indices into this study is necessary.

The Cu EF values are also presented in Table 4, according to the criterion of Zhang and Liu (2002), the sites at Kuala Kurau Town and Juru Industrial drainage had exceeded 1.5 value, suggesting that a significant portion of Cu is delivered from non-crustal materials or non-natural weathering processes (Feng et al. 1998) while other sampling sites may be entirely from crustal materials or natural weathering processes (Zhang and Liu 2002). Based on the Cu geochemical fractions (Table 3), this is true that Kuala Kurau Town and Juru Industrial drainage are dominated (>50%) by nonresistant fraction, thus, supporting that a significant portion of Cu is delivered from non-natural weathering processes or mostly by anthropogenic inputs. However, if other sampling sites may be entirely from natural sources, it is arguable since it is not supported by the Cu geochemical fractions (Table 4) for other sampling sites. Also, only Kuala Kurau Town had EF value greater than 2, suggesting various degrees of metal enrichment while other sites are considered as deficiency to minimal metal enrichment, according to the recommendation of Han et al. (2006). Interpretation by Acevedo-Figueroa et al. (2006) on the EF values suggests that sites at J.Raja Permaisuri Bainon, Kuala Kurau Town, Kuala Muda, Kuala Juru and Juru Industrial drainage are classified as “no-enrichment to minor enrichment” ($1 < EF < 3$) while other sites are considered as “no enrichment” at all ($EF < 1.0$). Therefore, the EF assessments based on Zhang and Liu (2002) can better classify the Cu contamination on the 18 sampling sites in the present study while the criterion of Acevedo-Figueroa et al. (2006) needs further modification to suit the Cu levels in the tropical sediment.

The Pb EF values are also presented in Table 5, according to the criterion of Zhang and Liu (2002), all the 18 sampling sites had exceeded 1.5 value, suggesting that a significant portion of Cu is delivered from non-crustal materials or non-natural weathering processes (Feng et al. 1998). This estimation is partly supported by the nonresistant fractions of Pb (Table 5) but the total Pb concentration is also contributed by the resistant or natural weathering sources as found in the present study based on geochemical fractions (Table 5). Therefore, the criterion of Zhang and Liu (2002) seems not to be applicable for the Malaysian Pb sediment and needs further revision and testing. Quite similarly, all the sampling sites except for Hutan Melintang site, had all EF values greater than 2, suggesting various degrees of metal enrichment, according to the recommendation by Han et al. (2006). The various degrees of metal enrichment are quite ambiguous and unclear. They can be due to both anthropogenic and natural sources. Hence, the criterion of Han et al. (2006) cannot assess the Pb contamination based on the present study. Lastly, the interpretation by Acevedo-Figueroa et al. (2006) on the EF values suggests that sites at Kuala Kurau Town is classified as “moderately severe enrichment” ($EF = 5–10$), sites at Pelabuhan Hutan Melintang, Bagan Datoh, J. Raja Permaisuri Bainon, Kuala Muda, and Juru Industrial drainage are classified as “is moderate enrichment” ($EF = 3–5$) while other sites are classified as “minor enrichment” ($1 < EF < 3$).

The Zn EF values are also presented in Table 6, according to the criterion of Zhang and Liu (2002), sites at Kuala Kurau Town, Kuala Juru Jetty, and Juru Industrial drainage had exceeded 1.5 value, suggesting that a significant portion of Cu is delivered from non-crustal materials or non-natural weathering processes (Feng et al. 1998). This estimation is partly supported by the nonresistant fractions of Zn (Table 6) but Juru Industrial drainage seems not to be dominated by nonresistant fraction (<50%) based on geochemical fractions (Table 6). The assessment of Zhang and Liu (2002) is well supported by the recommendation of Han et al. (2006) in which the above three sites exceeded 2.0 assessment criterion value. Lastly, the interpretation by Acevedo-Figueroa et al. (2006) on the EF values suggests that sites at Kampung Kuantan and Kuala Kurau Town are classified as “no enrichment to minor enrichment” ($EF = 1–3$) while sites at Kuala Juru Jetty and Juru Industrial drainage are classified as “moderate enrichment” ($EF = 3–5$). Since the assessment of Acevedo-Figueroa et al. (2006) can better classify the Pb and Zn contamination into different degrees of enrichment, it is the best

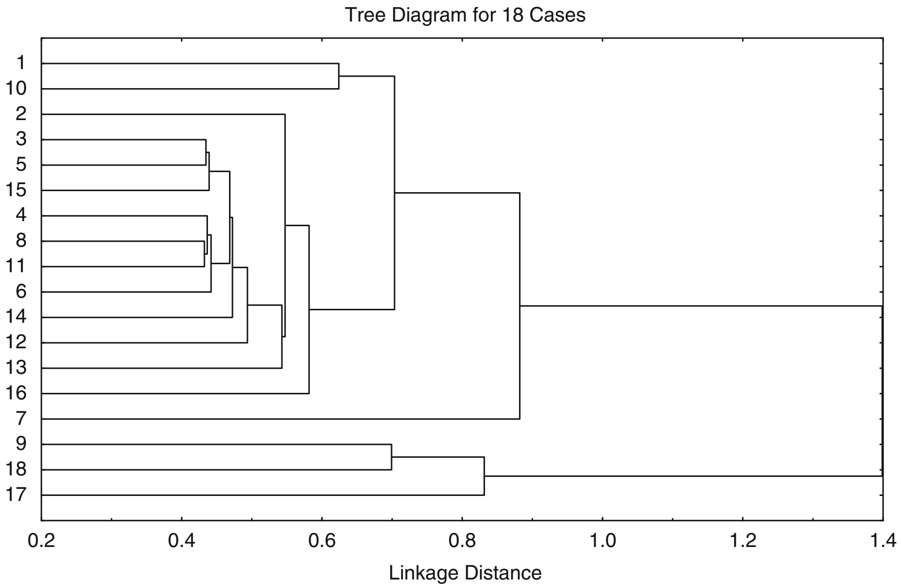

Fig. 2 Cluster analysis based on single linkage Euclidean distances, on the geochemical fractions of Cu, Pb, and Zn concentrations in the sediments collected from 18 sampling sites, based on $\log_{10}(\text{mean} + 1)$ transformed data of first three geochemical fractions (EFLE, acid-reducible and oxidisable-organic), ratios of nonresistant to resistant fractions and total concentrations of the Cu, Pb, and Zn. Note: Sampling site numbers follow those in Table 3

among the three criteria used so far regardless of some literature that all EF might represent the actual contamination level in the sediment (Groengroeft et al. 1998) and is a good tool to differentiate the metal source between anthropogenic and naturally occurring (Morillo et al. 2004; Selvaraj et al. 2004; Valdés et al. 2005).

Based on the Igeo classification in Table 2, for Cu Igeo indices (Table 4), all sampling sites had Igeo indices <0, indicating "unpolluted" condition except for Kuala Kurau Town (1.32; moderately polluted), Juru Industrial drainage (0.34; unpolluted to moderately polluted). For Pb Igeo indices (Table 5), all sampling sites had Igeo indices <1, indicating "unpolluted to moderately polluted" condition except for Kuala Kurau Town (2.39; moderately to strongly polluted), Juru Industrial drainage (1.44; moderately polluted) and Bagan Datoh (1.36; moderately polluted). For Zn Igeo indices (Table 6), all sampling sites had Igeo indices <1, indicating "unpolluted to moderately polluted" condition except for Kuala Kurau Town (1.17; moderately polluted), Kuala Juru Jetty (0.74; unpolluted to moderately polluted) and Juru Industrial drainage (1.35; moderately polluted).

Finally, based on the cluster analysis in Fig. 2, it can be summarized that sites at Kuala Kurau Town, Kuala Juru Jetty, and Juru Industrial drainage are grouped into a same sub-cluster, indicating that these two sampling sites (freshwater sediments) received more contamination of Cu, Pb, and Zn as shown in Tables 3 and 4 since these sites also had higher (>50%) nonresistant fractions of these metals than the resistant ones. These localized elevated metal concentrations could be related to point source discharges related to rapid urbanization and industrial development in Kuala Kurau Town and Juru adjacent areas, respectively. The Juru estuary was previously reported to have been polluted by heavy metals (Yap et al. 2002b; Yap et al. 2003). Therefore, the present data also confirmed continued point source of industrial discharge into the Juru river basin. The rest of the sites are clustered differently, indicating lesser contamination by Cu, Pb, and Zn. Therefore, the dendrogram based on cluster analysis supports the findings by using the assessment of geochemical study, EF, Igeo, and SQVs.

Conclusion

From the present study, it is evidently shown that sampling sites at Kuala Kurau Town and Juru Industrial drainage at Juru Industrial Area, were contaminated by Cu, Pb, and Zn based on ratios of nonresistant to resistant geochemical fractions, EF, Igeo and SQVs. However, caution should be exercised that the interpretation can only become better provided the ratios, indices and SQVs are combined besides examining the metal concentrations alone. This is due to the fact that not all the established indices are applicable and, to a certain extent, some of them should be further revised and improved to suit a different metal. An index established from other ecoregions could be useful for one metal but not necessarily for other metals. Thus, further improvement and revision on the geochemical indices are necessary based on Malaysian sediment data.

Acknowledgements The authors wish to acknowledge the financial support provided through the Research University Grant Scheme (RUGS) [Vot no: 91986] by University Putra Malaysia.

References

Acevedo-Figueroa, D., Jimenez, B. D., & Rodríguez-Sierra, C. J. (2006). Trace metals in sediments of two estuarine lagoons from Puerto Rico. *Environmental Pollution, 141*, 336–342.

Alagarsamy, R. (2006). Distribution and seasonal variation of trace metals in surface sediments of the Mandovi estuary west coast of India. *Estuarine Coastal and Shelf Science, 67*, 333–339.

Amin, B., Ismail, A., Arshad, A., Yap, C. K., & Kamarudin, M. S. (2009). Anthropogenic impacts on heavy metal concentrations in the coastal sediments of Dumai, Indonesia. *Environmental Monitoring and Assessment, 148*(1–4), 291–305.

Badri, M. A., & Aston, S. R. (1983). Observation on heavy metal geochemical associations in polluted and non-polluted estuarine sediments. *Environmental Pollution (Series B), 6*, 181–193.

Chapman, P. M., Patrick, J. A., & Vigers, G. A. (1999). Development of sediment quality values for Hong Kong special administrative region: A possible model for other jurisdictions. *Marine Pollution Bulletin, 38*, 161–169.

Chen, C. W., Kao, C. M., Chen, C. F., & Dong, C. D. (2007). Distribution and accumulation of heavy metals in sediments of Kaoshiung Harbor, Taiwan. *Chemosphere, 66*, 1431–1440.

Cheung, K. C., Poon, B. H. T., Lan, C. Y., & Wong, M. H. (2003). Assessment of metal and nutrient concentrations in river water and sediment collected from the cities in the Pearl River Delta, South China. *Chemosphere, 52*(9), 1431–1440.

Chloe, W. Y. T., Carman, C. I., Zhang, G., Paul, K. S. S., Qian, P., & Xiang-dong, L. (2008). The spatial and temporal distribution of heavy metals in sediments of Victoria Harbour, Hong Kong. *Marine Pollution Bulletin, 57*, 816–825.

Cuong, D. T., & Obbard, N. (2006). Metal speciation in coastal marine sediments from Singapore using a modified BCR sequential extraction procedure. *Applied Geochemistry, 21*, 1335–1346.

Cuong, D. T., Bayen, S., Wurl, O., Subramanian, K., Wong, K. K. S., Sivasothi, N., et al. (2005). Heavy metal contamination in mangrove habitats of Singapore. *Marine Pollution Bulletin, 50*, 1713–1744.

Daskalakis, K. K., & O'Connor, T. P. (1995). Normalization and elemental sediment contamination in the coastal United States. *Environmental Science & Technology, 29*, 47–477.

Ergin, M., Saydam, C., Basturk, O., Erdem, E., & Yoruk, R. (1991). Heavy metal concentrations in surface sediments from the two coastal inlets (Golden Horn Estuary and Izmit Bay) of the northeastern Sea of Marmara. *Chemical Geology, 91*, 269–285.

Feng, H., Cochran, J. K., Lwiza, H., Brownawell, B., & Hirschberg, D. J. (1998). Distribution of heavy metal and PCB contaminants in the sediments of an urban estuary: The Hudson River. *Marine Environmental Research, 45*, 69–88.

Gismera, M. J., Lacal, J., da Silva, P., Garcia, R., Sevilla, M. T., & Procopio, J. R. (2004). Study of metal fractionation in river sediments. A comparison between kinetic and Sequential Extraction procedure. *Environmental Pollution, 127*, 175–182.

Groengroeft, A., Jaehnig, U., Miehlich, G., Lueschow, R., Maass, V., & Stachel, B. (1998). Distribution of metals in sediments of the Elbe Estuary in 1994. *Water Science and Technology, 37*, 109–116.

Han, Y. M., Du, P. X., Cao, J. J., & Posmentier, E. S. (2006). Multivariate analysis of heavy metal contamination in urban dusts of Xi'an, Central China. *The Science of the Total Environment, 355*, 176–186.

Ismail, A. (1993). Heavy metal concentration in sediment of Bintulu, Malaysia. *Marine Pollution Bulletin, 26*, 706–707.

Ismail, A., Badri, M. A., & Ramlan, M. N. (1993). The background levels of heavy metals concentration in sediments of the west coast of Peninsular Malaysia. *The Science of the Total Environment, Supplement, 1993*(suppl), 315–323.

Ismail, A., Yap, C. K., & Chan, F. F. (2004). Concentrations of Cd, Cu and Zn in sediments collected from urban lakes at Kelana Jaya, Peninsular Malaysia. *Wetland Science, 2*(4), 248–258.

Li, Q. S., Wu, Z. F., Chu, B., Zhang, N., Czi, S. S., & Fang, J. H. (2007). Heavy metals in the coastal wetland sediments of the Pearl River Estuary, China. *Environmental Pollution, 149*, 158–164.

Lim, P. E., & Kiu, M. Y. (1995). Determination and speciation of heavy metals in sediments of the Juru River, Penang, Malaysia. *Environmental Monitoring and Assessment, 35*, 85–95.

MacDonald, D. D., Ingersoll, C. G., & Berger, T. A. (2000). Development and evaluation of consensus-based sediment quality guidelines for freshwater ecosystems.

Archives of Environmental Contamination and Toxicology, 39, 20–31.

Martin, J. M., & Whitfield, M. (1983). The significance of the river input of chemical elements to the ocean. In C. S. Wong, E. Boyle, K. W. Brul, J. D. Burton, & E. D. Goldberg (Eds.), *Trace metals in sea water* (pp. 265–296). New York: Plenum.

Mat, I., Maah, M. J., & Yahya, M. S. (1994). The status of trace metal pollution in inter-tidal sediments from the Straits of Tebrau, Malaysia. *Jurnal Sains, 2*(2), 221–228.

Morillo, J., Usero, J., & Gracia, I. (2004). Heavy metal distribution in marine sediments from the southwest coast of Spain. *Chemosphere, 55*, 431–442.

Müller, G. (1981). Die Schwermetallbelastung der sedimente des Neckars und seiner Neben.usse: eine Bestandsaufnahme. *Chemical Zeitung, 105*, 157–164.

Mucha, A. P., Vasconcelos, M. T. S. D., & Bordalo, A. A. (2003). Macrobenthic community in the Doura estuary: Relations with trace metals and natural sediment characteristics. *Environmental Pollution, 121*, 169–180.

Praveena, S. M., Radojevic, M., Abdullah, M. H., & Aris, A. Z. (2008). Application of sediment quality guidelines in the assessment of mangrove surface sediment in Mengkabong Lagoon, Sabah, Malaysia. *Iranian Journal of Environmental Health, and Science Engineering, 5*(1), 35–42.

Ramirez, M., Massolo, S., Frache, S., & Correa, J. A. (2005). Metal speciation and environmental impact on sandy beaches due to El Salvador copper mine, Chile. *Marine Pollution Bulletin, 50*, 62–72.

Reddy, M. S., Basha, S., Kumar, V. G. S., Joshi, H. V., & Ramachandraiah, G. (2004). Distribution, enrichment and accumulation of heavy metals in coastal sediments of Alang-Sosiya ship scrapping yard, India. *Marine Pollution Bulletin, 48*, 1055–1059.

Salomons, W., & Forstner, U. (1984). *Metals in the hydrocycle* (p 349). Berlin: Springer.

Schi, K. C., & Weisberg, S. B. (1999). Iron as a reference element for determining trace metal enrichment in Southern California coast shelf sediments. *Marine Environmental Research, 48*, 161–176.

Selvaraj, K., Mohan, V. R., & Szefer, P. (2004). Evaluation of metal contamination in coastal sediments of the Bay of Bengal, India: Geochemical and statistical approaches. *Marine Pollution Bulletin, 49*, 174–185.

Shen, Y. (1992). Distribution of contents of Fe_2O_3, Al_2O_3 and Ti_2O and its relationship to surface sediments in the Xiamen Harbour. *Journal of Oceanography in Taiwan Strait, 11*, 146–151. (in Chinese with English abstract).

Takarina, N. D., David, R. B., & Micheal, J. R. (2004). Speciation of heavy metal in coastal sediments of Semarang, Indonesia. *Marine Pollution Bulletin, 49*, 854–874.

Turekian, K. K., & Wedepohl, K. H. (1961). Distribution of the elements in some major units of the earth's crust. *Bulletin of Geological Society of America, 72*, 175–192.

Valdés, J., Vargas, G., Sifeddine, A., Ortlieb, L., & Guinez, M. (2005). Distribution and enrichment evaluation of heavy metals in Mejillones Bay (23°S), Northern Chile: Geochemical and statistical approach. *Marine Pollution Bulletin, 50*, 1558–1568.

Wedepohl, K. H. (1995). The composition of the continental crust. *Geochimica et Cosmochimica Acta, 59*(7), 1217–1232.

Wood, A. K., Ahmad, Z., Shazili, N. A., Yaakob, R., & Carpenter, R. (1997). Geochemistry of sediments in Johore Strait between Malaysia and Singapore. *Continental Shelf Research, 10*, 1207–1228

Yap, C. K., Ismail, A., & Tan, S. G. (2002a). Correlations between speciation of Cd, Cu, Pb and Zn in sediment and their concentrations in soft tissue of green-lipped mussel Perna viridis from the west coast of Peninsular Malaysia. *Environment International, 28*, 117–126.

Yap, C. K., Ismail, A., & Tan, S. G. (2002b). Concentrations of Cu and Pb in the offshore and intertidal sediments of the west coast of Peninsular Malaysia. *Environment International, 28*, 467–479.

Yap, C. K., Ismail, A., & Tan, S. G. (2003). Cd and Zn concentrations in the straits of Malacca and intertidal sediments of the west coast of Peninsular Malaysia. *Marine Pollution Bulletin, 46*, 1341–1358.

Yap, C. K., Choh, M. S., Edward, F. B., Ismail, A., & Tan, S. G. (2006a). Comparison of heavy metal concentrations in surface sediment of Tanjung Piai wetland with other sites receiving anthropogenic inputs along the southwestern coast of Peninsular Malaysia. *Wetland Science, 4*(1), 48–57.

Yap, C. K., Ismail, A., Pang, B. H., Yeow, K. L., Tan, S. G., & Siraj, S. S. (2006b). Elevated heavy metal concentrations in surface sediments collected from the drainages of the Sri Serdang Industrial Area, Malaysia. *Malaysian Applied Biology, 35*(2), 35–40.

Yap, C. K., Ismail, A., Low, C. H., & Tan, S. G. (2007a). Interpretation of Cu and Zn contamination in the aquatic environment of Peninsular Malaysia with special reference to a polluted river, Sepang River. *Wetland Science, 5*(4), 311–321.

Yap, C. K., Pang, B. H., Fairuz, M. S., Hoo, Y. I., & Tan, S. G. (2007b). Heavy metal (Cd, Cu, Ni, Pb and Zn) pollution in surface sediments collected from drainages receiving different anthropogenic sources from Peninsular Malaysia. *Wetland Science, 5*(2), 97–104.

Yap, C. K., Cheng, W. H., & Tan, S. G. (2008a). Comparative studies of concentrations of Cu and Zn in the surface intertidal sediments collected from east, south and west coasts of Peninsular Malaysia. *Asian Journal of Water, Environment and Pollution, 5*(2), 23–29.

Yap, C. K., Fairuz, M. S., & Cheng, W. H. (2008b). How elevated of Cd, Cu and Pb levels in the surface sediments collected from drainage receiving metal industrial effluents? Comparison with metal industrial drainage and intertidal sediments in Selangor, Malaysia. *Asian Journal of Microbiology, Biotechnology and Environmental Sciences, 10*(2), 385–391.

Yap, C. K., Fairuz, M. S., Cheng, W. H., & Tan, S. G. (2008c). Distribution of Ni and Zn in the surface sediments collected from drainages and intertidal area in Selangor. *Pertanika Journal of Tropical Agricultural Science, 31*(1), 79–90.

Yap, C. K., Yazdani, M., & Tan, S. G. (2009). The importance of the identification of point sources in a river receiving industrial metal effluents at the Serdang Industrial Area, Selangor. *Journal of Sustainability Science and Management, 4*(2), 18–26.

Zhang, J., & Liu, C. L. (2002). Riverine composition and estuarine geochemistry of particulate metals in China—Weathering features, anthropogenic impact and chemical fluxes. *Estuarine, Coastal and Shelf Science, 54*, 1051–1070.

Zhang, L., Ye, X., Feng, H., Jing, Y., Ouyang, T., Yu, X., et al. (2007). Heavy metal contamination in western Xiamen Bay sediments and its vicinity, China. *Marine Pollution Bulletin, 54*, 974–982.

Zhang, W., Feng, H., Chang, J., Qu, J., Xie, H., & Yu, X. (2009). Heavy metal contamination in surface sediments of Yangtze River intertidal zone: An assessment from different indexes. *Environmental Pollution, 157*, 1533–1543.